Laboratory Name: Sandia National Labs/NM
B&R Code: KC020103

FWP: Atomic Scale Surface Phenomena

FWP Number: SCW 93219

Program Scope: Our goal is to develop an atomic-scale understanding of physical and chemical processes at surfaces and interfaces in three major thrust areas: (1) adhesion and wetting, (2) localized corrosion, and (3) surface nanostructure formation. Our approach is to combine state-of-the-art experimental probes with modern theoretical techniques to produce the scientific understanding necessary to predict and control macroscopic materials properties.

Major Program Achievements (over duration of support): We invented the interfacial force microscope (IFM), the world's first (and only) scanning probe instrument that can measure normal and lateral forces simultaneously throughout the entire attractive/repulsive interaction regime. With the IFM we have achieved an understanding of two-stage plasticity in nanoscale contacts, yield strength softening by surface defects, and interphase mechanical properties in nanocomposite materials. We identified distinct physical and chemical contributions to friction for self-assembled alkanethiol monolayers in dynamic contact, mechanisms of energy dissipation, and environmental degradation processes. We discovered that when aluminum is exposed to water, nanoscale open pores develop at the oxide surface and these pores are linked to aluminum pitting in the presence of aggressive species. Through careful control of the electrochemical parameters, we showed that these pores initiate as voids at the oxide/metal interface and grow towards the surface, with the highest concentrations developing at high-curvature surfaces. We discovered important new mechanisms for dynamic surface processes including exchange- and vacancy-mediated surface diffusion, and a remarkable self-assembly process for Pb atoms on Cu surfaces. We acquired an atomistic understanding of spontaneous stripe formation, structural phase transitions, and oxygen-induced etching on Si surfaces. We developed new surface probes including the atom-tracking scanning tunneling microscope, the pulsed-laser atom probe, LEEM I-V analysis, and a first-principles code for investigations of atoms and defects on surfaces.

Program Impact: The IFM is finding broad application in the international research community (17 IFMs in US/Canada). Our approach of using engineered defects in controlled oxides to investigate the early stages of pitting has allowed us to address outstanding questions in corrosion science in a clean and controlled manner. It has also influenced the experimental design used by other groups within the corrosion community. Surface nanostructure research is providing new insights on the microscopic origins of crystal and thin-film growth, surface phase transitions, and surface self-assembly. Adhesion/wetting and surface nanostructure research has been highlighted in *Science, Nature, Physics Today, Popular Science, Popular Mechanics, Wall Street Journal, MRS Bulletin, etc*.

Interactions: Princeton U., U. Houston, U. Minnesota, U. Pittsburgh, Carnegie Mellon, U. Wisconsin at Madison, South Dakota School of Mining and Tech., U. Texas at Austin, U. Western Ontario, Ohio State, North Carolina State, Penn State, Rutgers U., U. of Twente (Netherlands), U. Texas at El Paso, New Mexico State, U. New Mexico, Brookhaven National Labs, Los Alamos National Lab, IBM Yorktown Heights, KFA-Jülich.

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):
Five BES/MS Awards (Houston, Kellogg (2), Feibelman (2), Swartzentruber); 1996 AVS Welch Award (Feibelman); 1997 AVS Peter Mark Award (Swartzentruber); DOE OER's Young Scientist Award (Swartzentruber); 2001 Materials Research Society Medal (Bartelt), 2004 Stanley E. Harrison Faculty Award, OSU (Buchheit); Fellows of the AAAS (Houston), APS (Houston, Feibelman, Kellogg, Bartelt), AVS (Houston, Feibelman, Kellogg, Swartzentruber), numerous editorial boards (PRL, JVST, Prog. in Surf Sci.), conference and symposium organizers, national and international scientific committee positions, and invited talks (>20 since 2001).

Personnel Commitments for FY2005 to Nearest +/-10%: (Staff) G. L. Kellogg 50%, J. E. Houston 40%, B. S. Swartzentruber 40%, P. J. Feibelman 20%, W. L. Smith 40%, N. Missert 50%, K. R. Zavadil 40%, L. Serna 40%, J. P. Sullivan 20%, P. Kotula10%, N. Simmons (technologist) 10%, N. Vasiljevic (post-doc) 70%, D. Elswick (NSCU grad student) 100%, Y. Kim (OSU grad student) 100%

Authorized Budget (BA) for FY03, FY04, FY05:
FY03 BA: $1,620 K **FY04 BA:** $1,610 K **FY05 BA:** $1,521 K

Laboratory Name: Sandia National Laboratories
B&R Code: KC020202

FWP: Quantum Electronic Phenomena and Structures
FWP Number: SCW 93220

Program Scope: This project explores growth and fabrication of nanoelectronic structures in order to manipulate quantum electronic and optical properties. The three subtasks th are to study the role of interactions in low dimensional systems, to study the growth and properties of quantum dot arrays and vapor-liquid-solid (VLS) 1D structures, and to study transport and coherence in artificially patterned 1D quantum wires and antidot arrays.

Major Program Achievements (over duration of support):

Percolation in low density 2D electrons: The density dependence of the conductivity has been experimentally and theoretically analyzed in the context of density inhomogeneities and percolation. The data fit very well to a model of percolation scaling and show that percolation phenomena are important in the low density limit for the 2D metal-insulator transition.

Electron-hole bilayers: Techniques to fabricate high mobility and low density 2D electrons and holes have been extended to bilayer systems. We have demonstrated the first ever undoped electron-electron, hole-hole and electron-hole bilayers using field-effect gating techniques.

III-nitride nanowire arrays: Dense, highly aligned arrays of sharp-tipped, single-crystalline GaN nanowires have been synthesized on sapphire substrates without the use of a template. Results indicate that the alignment and size are strong functions of substrate orientation and catalyst concentration, while the structural, electrical, and optical properties of the nanowires depend strongly on the growth temperature.

Quantum dot superlattices for Bloch oscillations: High quality and large area quantum dot superlattices are defined using the interferometric lithograph technique (with Steve Brueck, UNM). In I-V measurements we have observed negative differential conductance whose origin is possibly due to radiation from intersubband transitions. In a novel setup of pure DC magnetoresistance measurement, an anomalous resistance spike was observed; one possible origin is due to coupling radiation and edge magnetoplasmons.

Tunneling in 1D wires: Vertically coupled quantum wires are fabricated. The density in each wire can be independently controlled, and separate contacts can be made to each quantum wire. Tunneling spectroscopy between the quantum wires is measured and strong signatures of each 1D subband are visible. Non-interacting models capture some of the basic features observed, but additional rich structure is present.

Program impact: Experiment and theory are combined to discover and understand some of the most exciting physics in compound semiconductor research today. This is achieved by understanding how to utilize nature to grow unique structures and then identify the physics of these structures. Advances in materials growth (high mobility GaAs/AlGaAs, direct nanostructure systhesis), advanced electronic measurements (tunneling, low density transport) and processing (vertical quantum wires, quantum dot arrays) and optics all contribute to our understanding of quantum phenomena in low dimensional systems.

Interactions: Collaborations have been established with Princeton (D. Tsui), Caltech (J. Eisenstein), Univ. of Maryland (S. Das Sarma), NRL (A. Efros), Univ. of Bath (P. Russell and J. Knight), SUNY Buffalo (J. Bird), Rice University (R. Du) and the National High Magnetic Field Laboratory. This project has enhanced collaborations between Sandia National Labs, Los Alamos National Labs and the Univ. of New Mexico.

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):
Chairs of the 16th International Conference on the Electronic Properties of 2D Systems (Simmons, Lilly); V. I. Klimov, LANL Laboratory Fellow (2003); J. A. Simmons, Fellow of the American Physical Society (2002); Invited presentations at the APS March meeting (Reno, 2005; Lilly, 2004; Klimov, 2003; Taylor, 2003), EPQHS 2005 (Pan, Lilly), SPIE, 2003 MRS Fall meeting; numerous invited seminars and colloquia.

Personnel Commitments for FY2005 to Nearest +/- 10%:
M. P. Lilly (30%), Wei Pan (30%), Ken Lyo (50%), John Reno (20%), Victor Klimov (30%), Toni Taylor (20%), S. Brueck (20%), George Wang (30%)

Authorized Budget (BA) for FY06, FY07, FY08:
FY03 BA $920K **FY04 BA** $990K **FY05 BA** $1,170K (includes Q Dot Arrays subtask)

Laboratory Name: Sandia National Labs/NM
B&R Code: KC020102

FWP: Mechanics of Small Length Scales

FWP Number: SCW 93221

Program Scope: Understanding mechanical response at the nanoscale to obtain atom-level control over synthesis and properties of nanostructured materials with designed mechanical functionality. Investigates (i) Synthesis, dissipation, and elastoplastic response of nanocrystalline aggregates, nanoelectromechanical structures (NEMS), quantum dots and wires, and (ii) Detailed nanoscale mechanical interaction behavior underlying mesoscale deformation, material transport, collective phenomena, and phase stability.

Major Program Achievements (over duration of support):

Nanostructured Materials: **Direct in situ TEM evidence for grain-boundary deformation processes occurring simultaneously with dislocation propagation in 10 nm grain-size Ni, having very high strength (4-5 GPa) consistent with such deformation. **Exceptional strengthening of metals by very high densities of either precipitates or cavities (1-10 nm size); strength accounted for by Orowan hardening. Quantitative understanding of tensile and compressive residual film stresses via unique selective growth approach.

Theory of Microstructures: ** First physically-based model for nucleation of recrystallization from deformation substructure. **New molecular dynamics method for computing the mobility of flat grain boundaries with/without solute present. **First characterization of abnormal growth due to texture gradients. **First proof that genetic algorithms can beat simulated annealing for structure optimization. **New scaling theory for 2D/3D grain boundary networks proving R-curve behavior is intrinsic to polycrystals.

Advanced Growth: **Developed novel real-time diagnostics for thin film growth: energy dispersive x-ray reflectivity, multibeam optical stress sensor, and light scattering spectroscopy. **Quantitative understanding of morphological evolution of dense arrays of heteroepitaxial SiGe quantum dots. **Kinetically-controlled heteroepitaxial self-assembly to produce complex functional nanostructures with precision placement. **Characterized fracture energetics and slip processes in strained III-nitride heterostructures.

Plasma Crystals: **First measurements of the ion wakefield induced binding potential in 3D crystals. **Developed new laser techniques to determine the sheath electric field. **First measurements of the electric field distribution on particles immersed in a plasma to validate historical models of Langmuir probe effects. **Developed a new analysis technique to directly determine the particle-particle interactions potential in 2D plasma crystals. **Used curved electrode structures to modify the plasma potential.

Program impact: We developed methods to produce, characterize, and theoretically analyze unusually strong metals via nanostructuring, providing a scientific basis for designing new mechanical properties. Our new computational methods with enhanced efficiencies for evolving realistic micro/nanostructures reveal new behaviors and provide better understanding for tailoring of materials. Using novel real-time diagnostics for film growth we developed a deep understanding of the energetics/kinetics of heteroepitaxial self-assembly to create structures pertinent to nanologic. Our novel plasma diagnostics and approaches provide basic insights into the long range, collective interactions between microdust particles.

Interactions: AFRL, Baylor, Brazilian LNLS, BNL, Brown, CMU, DOE/BES CompMatSciNet, T.U. Eindhoven, Harvard, Hong Kong Polytech, U. Houston, UIUC, IBM, U. Iowa, King's College, MPI, MIT, Mich. St. U., NRL, UNM, Scherrer Inst., Pitt, UVa. *(Names of collaborators provided upon request)*

Recognitions, Honors and Awards: Floro: MRS BoD 02-04, MRS Public Outreach Chr, MRS Mtg Chr 01, BES Outstand Sust Rsrch Award 94. **Follstaedt**: R&D100 04, BES Mat Sci Award 91. **Hebner**: AVS Plasma Prize, AVS Fellow. **Holm**: NMAB 04-07, TMS BoD 04-07, THERMEC 06 Intl Sci Comm, PRICM5 org 04, NWU John Dorn Lecturer (2005), U. Mich. Faculty Grant 04, Fellow ASM 02.

Invited conference / academic presentations since 1995: 117 Archival publications since 1995: 120

Personnel Commitments for FY2005 to Nearest +/- 10%: Barnat 30%, Farrow 100%, Floro 90%, Follstaedt 30%, Hearne 20%, Hebner 10%, Holm 50%, Knapp 20%

Authorized Budget (BA) for FY03, FY04, FY05:

FY03 BA: $1,192K **FY04 BA:** $1,192K **FY05 BA:** $1,030K

Laboratory Name: Sandia National Labs / NM
B&R Code: KC 020103

FWP and subtasks under FWP:
FWP: Novel Electronic Materials. Subtasks: [1] Luminescence in Wide-Bandgap Semiconductors; [2] Atomic Processes and Defects in Wide-Bandgap Semiconductors; [3] Field-Structured Composites; [4] Dipolar Nanocomposites; [5] Complex and Cooperative Phenomena in Disordered Ferroelectrics and Dielectrics.

FWP Number: SCW 93222

Program Scope:
We are achieving new fundamental understanding of how atomic structure and nanostructure influence the electrical, optoelectronic, ferroelectric, and magnetic properties of novel and advanced materials and are developing new methods of nanoscale manipulation for property enhancement. Subtasks [1] and [2] are elucidating defects and compositional fluctuations in GaN/GaInN that alter carrier transport, recombination paths, and luminescence mechanisms; [3] and [4] are showing how self-assembly in triaxial magnetic and electric fields can produce tailored composites with highly attractive magnetic, transport, and mechanical properties; and [5] is revealing fundamentals of cooperative phenomena in disordered quantum ferroelectrics, thereby providing a basis for property optimization.

Major Program Achievements (over duration of support):

- Our work on III-nitrides is enabled by the development of a unique range of capabilities encompassing growth, experimental characterization, ab-initio theory, and modeling. Key recent advances in growth include a new MOCVD reactor and the discovery that strain-relaxed growth doubles achievable In concentrations in InGaN. [1]
- An integrated effort including experiments, density-functional theory, and modeling yielded quantitative new understanding of the N vacancy (V_N) and N interstitial (N_I) in GaN. This understanding encompasses structure, diffusion, reactions with H and Mg dopants, and their technologically important consequences for p-type doping. Recent achievements include the prediction and experimental detection of the bound complex $MgHN_I$; measurement and prediction of the temperature-dependent diffusivity of N_I; and a predictive model of the device-performance-limiting interplay of dopants, H, and V_N during growth and annealing of p-type GaN. [2]
- To illuminate carrier behavior underlying luminescence in III-nitride structures, we investigated the quantum-confined Stark effect in InGaN/GaN single quantum wells. This led to a new model of hole-localization which accounts for anomalous Stark shifts observed in these highly strained structures. Further, we used a new pulse-recovery technique to determine the transport properties of minority-carrier holes in GaN. [1]
- We discovered that time-varying, triaxial magnetic fields can be used to synthesize a variety of new particle composites with exceptional properties. Some of these materials enable chemical, pressure, and thermal sensing with extraordinary resolution, *e.g.*, detection of organic vapors at 300 ng/ml. Others exhibit magnetostriction as large as 1% (very attractive for actuators); magnetoresistance extending to 10,000,000 % in magnetic fields as low as 0.1 T; and superparamagnetism with susceptibilities up to 180 (MKS). These studies include new theoretical models describing the field-influenced dynamics of stirring, agglomeration, and ordering. [3,4]
- In studies of compositionally disordered ABO_3 ferroelectrics, we discovered and understood a crossover to a relaxor state with only short-range order, confirmed by the interplay of pressure (favoring the relaxor) and a biasing electric field (promoting long-range order). Our studies of this general phenomenon showed that the relaxor is the ground state at reduced volume; provided insights into static and dynamic properties; and enabled fundamental interpretation in terms of the correlation length for dipolar interactions among polar nano-regions. [5]

Program impact:
Basic knowledge supporting solid-state lighting with a potential for US energy savings up to $ 30 billion/year [1,2]. Four patents awarded on new composites with two in preparation [3,4]. Breakthrough in understanding relaxors, a premier current topic in ferroelectricity with large technological import. [5]

Interactions: 7 universities & research institutions; 2 semiconductor companies.

Recognitions, Honors and Awards (at least partly attributable to support under this FWP):
Many invited papers, 3 in 05-06; many symposia & conferences chairs, 2 in 05-07; 3 editorial & conference boards.

Personnel Commitments for FY2005 to Nearest +/- 10%:
Myers 30% [2]; Lee 25%, Crawford 15%, Follstaedt 10%, Kaplar 10% [1]; Fleming 10%, Koleske 20% [1,2]; Wright 10%, Wampler 10%, Wixom 40% [2]; Martin 40%, Anderson 10%, Huber 10%, [3,4]; Venturini 20% [4,5]; Samara 20% [5].

Authorized Budget (BA) for FY03, FY04, FY05:
FY03 BA $1,049K FY04 BA $1,484K FY05 $1416K

Laboratory Name: Sandia National Labs/NM
B&R Code: KC020301

FWP and possible subtask under FWP: Molecular Nanocomposites
FWP Number: SCW 93223

Program Scope: This project develops a fundamental understanding of the principles that govern the formation and function of novel nanoscale and nanocomposite materials. The scope of scientific issues being addressed include: design and synthesis of complex molecular precursors, nanoclusters and nanoparticles; formation of robust 2- and 3-D ordered nanocomposites; understanding of self-assembly mechanisms and the properties derived from the nanoscale (e.g., transport, electronic, interfacial chemistry, etc.).

Major Program Achievements (over duration of support): Nanocluster synthesis and characterization: Developed first room T synthesis of nanocrystalline Si and Ge in high yield with size tunable visible luminesence. Demonstrated: 1) the importance of alloying of non-magnetic atoms like Pt and Ag with Co, Ni, and Fe to achieve greater than bulk magnetic response and 2) cluster surface restructuring to create a white light nanophosphor using a single size semiconductor nanocrystal.

Nanocrystal synthesis: Demonstrated that the structure of precursors used in nanocrystal synthesis plays a vital role in nanocrystal morphology development and that molecular design of these precursors can be used to control morphology evolution (i.e., dots versus wires).

Molecular assemblies: Developed 1) new understanding of molecular organization in dynamic assemblies via synthetic lipids and surfactants that impart novel 2- and 3-D structure and 2) imaging and force measurement techniques enabling characterization of nanoscale chemistry and molecular dynamics in self-organized film and cell surfaces.

Nanostructural architecture and nanocomposites: Developed new approaches to hierarchical nanostructures using surfactant / lipid assemblies as structure directing agents in metal and metal oxide systems, including Pt and ZnO. Discovered a new nanofabrication approach, cell-directed assembly, in which living cells direct the formation of functional bio/nano interfaces allowing the development of 3D chemical gradients.

Self-assembly: Extended work on the formation of gold nanocrystal (NC) micelles and their self-assembly with soluble silica into 3-D gold NC/silica arrays to include: 1) semiconductor and magnetic NCs and 2) 2- or 3-D Au/silica NC arrays where silica is provided exclusively by silane moieties allowing greater control of NC spacing and, presumably, charge transport.

Nano-assembly characterization: Developed high-speed ^{1}H and ^{1}H-X nucleus NMR correlation method to probe interactions and domain sizes on the nanoscale in self-assembled and nanostructured materials.

Super-hydrophobic surfaces: Demonstrated how micro-to-milliscale roughness or patterning, along with a super-hydrophobic coating, can result in effective slip lengths comparable to the feature size.

Program impact: This program has stimulated research in the integration of molecular/biomolecular function with nanoscale structures, leading to additional funding from NIH, AFOSR, and ARO. New internal programs based on nanocluster materials were initiated in radiation detection, IR photovoltaics, catalysis, and taggants for homeland security. The controlled construction of nanoparticles allows exploration of very complex species and architectures to elucidate new and exploitable processes and physics. The new fundamental knowledge of super-hydrophobic surfaces will help in the design of effective drag reducing coatings.

Interactions: University: 30 profs./19 univs National Lab: LANL/LANL(LANSCE)/LBNL/LLNL

Recognitions, Honors and Awards: Publications in Science, Nature, Nano Letters, Physical Review Letters; JACSMRS Medal (Brinker '03); DOE's Lawrence Award (Brinker '02); MRS Graduate Student Gold Award (Baca '05); Advisory Board - NIH Center for Biomedical Research (Sasaki); MRS, AVS, and Pacifichem Symp. Organizer (Sasaki, Shreve); Editorial Board of *Nanotechnology* (Shelnutt); 3 BES awards

Personnel Commitments for FY2005 to Nearest +/- 10%: T. Boyle (30 %), T. Alam (30%), F. VanSwol (30%), C. J. Brinker (20%), J. Wilcoxon (30%), E. Venturini (20%), P. Provencio (20%), B. Abrams (20%), S. Thoma (10%), G. Galli (10%), A. Burns (20%), B. Bunker (20%), D. Sasaki (20%), C. J. Brinker (20%), J. Shelnutt (20%).

Authorized Budget (BA) for FY03, FY04, FY05:
FY03 BA: $1,688K **FY04 BA:** $1,668K **FY05 BA:** $1,595K

Laboratory Name: Sandia National Labs/NM
B&R Code: KC020105

FWP: Biointegration
FWP Number: SCW 93411

Program Scope: This project involves fundamental materials science studies at the intersection of biology, nanomaterials, and integrated systems. It represents the recent merger of two smaller projects: 1) Active Assembly of Dynamic and Adaptable Materials, which involves learning how to exploit active biomolecules such as motor proteins and microtubules to transport, assemble, and reconfigure nanomaterials, and 2) Artificially Structured Biocompatible Semiconductors, which involves learning how to develop semiconductor and photonic micro-devices for interrogating and manipulating biomaterials through the use of bio-compatible interfacial nano-materials.

Major Program Achievements (over duration of support):
Active Transport of Nanomaterials: Stabilized active proteins (motor proteins and microtubules) have been produced for use in artificial systems via both genetic engineering and chemical crosslinking. Monolayers containing the motor protein kinesin have been used to propel microtubule shuttles plus associated nanoparticle cargo within patterned microfluidic devices. Fundamental nanomechanics studies have been performed to understand cargo loading and unloading phenomena, as well as the formation of mobile particle-microtubule composites.

Artificial Microtubule Organizing Centers (AMOCs): 3D constructs consisting of a central functionalized particle, and oriented array of microtubules radiating out from the particle, and molecular motors plus associated cargo have been synthesized. Simulation codes have been developed to model how such constructs can be used to move and assemble nanomaterials via dynamic instability of the microtubules plus motor protein motion, driving experimental efforts to duplicate processes such as diatom assembly and the color changing system of the chameleon.

Biocompatible Semiconductors: III-V strained layer superlattices have been lithographically patterned to produce bio-cavity lasers that emit light at wavelengths ranging from 270 nm to over 10 microns. The surfaces of the bio-cavity laser have been functionalized with a wide range of dielectric overlayers and monolayers to protect the semiconductors from biological fluids and to provide biocompatible interfaces. Fundamental physics studies have been performed to determine how confined light in the laser cavity interacts with a wide range of biomaterials.

Program impact: The Active Assembly activity has demonstrated that energy-consuming protein machines can assemble and manipulate materials in new ways that are not limited by diffusion and energy minima constraints encountered in the classical self-assembly of nanomaterials. The project also highlights a new transport mechanism for moving nanomaterials in microfluidic systems that has impacted the development of “on-chip” analysis systems for Homeland Defense (DARPA). The Biocompatible Semiconductors project developed the Biocavity Laser, which has enormous potential for the on-chip interrogation of biomaterials and for the development of integrated Microsystems as tools for biomedical research funded by NIH.

Interactions: In addition to co-investigators H. Hess (U. Florida) and V. Vogel (now at ETH, Switzerland), the project has stimulated interactions with R. Haddon (UC Riverside), D. Bear, J. Oliver, and M. Keep (U. of New Mexico), and M. Gourley (NIH).

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):
2005 Research Prize, German Phillip Morris Foundation (Hess and Vogel)
Symposia Organizer/Chair for 2 MRS Meetings + 2 AAAS Meetings (Bachand and Liu)
Cover story in Advanced Functional Materials (1/04) + Invited Review article in Materials Today (in press)
Thirteen publications (3 invited) + 12 invited talks

Personnel Commitments for FY2004 to Nearest +/- 10%:
B. Bunker (35%), P. Gourley (50%), G. Bachand (30%), J. Liu (30%), G. Osbourn (50%), D. Sasaki (25%), H. Hess (20%), V. Vogel (25%)

Authorized Budget (BA) for FY03, FY04, FY05:
FY03 BA: $1,241 K **FY04 BA:** $1,241 K **FY05 BA:** $1,221 K